

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

Mo-6925/LeA 33,899

U.S. APPLICATION NO. (If known, see 37 CFR 1.5

10/031502  
To Be Assigned

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/EP00/06377

06 July 2000 (6.07.00)

19 July 1999 (19.07.99)

TITLE OF INVENTION

POLYCARBONATE AND MOLDED POLYCARBONATE ARTICLES

APPLICANT(S) FOR DO/EO/US KRATSCHMER, Silke; HUCKS, Uwe and BUNZEL, Lothar

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☒ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☒ is attached hereto.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:

Form PTO 1449 w/references

Abstract

21. ☒ The following fees are submitted:

**BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):**

Neither international preliminary examination fee (37 CFR 1.482)  
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO  
and International Search Report not prepared by the EPO or JPO. . . . . **\$1040.00**

International preliminary examination fee (37 CFR 1.482) not paid to  
USPTO but International Search Report prepared by the EPO or JPO . . . . . **\$890.00**

International preliminary examination fee (37 CFR 1.482) not paid to USPTO  
but international search fee (37 CFR 1.445(a)(2)) paid to USPTO . . . . . **\$740.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO  
but all claims did not satisfy provisions of PCT Article 33(1)-(4) . . . . . **\$710.00**

International preliminary examination fee (37 CFR 1.482) paid to USPTO  
and all claims satisfied provisions of PCT Article 33(1)-(4) . . . . . **\$100.00**

**ENTER APPROPRIATE BASIC FEE AMOUNT =**

**CALCULATIONS PTO USE ONLY**

\$ 890.00

Surcharge of **\$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$ 0.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	9 - 20 =	0	x <b>\$18.00</b>
Independent claims	1 - 3 =	0	x <b>\$84.00</b>

\$

\$ 0.00

MULTIPLE DEPENDENT CLAIM(S) (if applicable)	+ <b>\$280.00</b>
<b>TOTAL OF ABOVE CALCULATIONS =</b>	

\$ 0.00

\$ 0.00

\$ 890.00

☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above  
are reduced by 1/2.

\$

0.00

**SUBTOTAL =**

\$ 890.00

Processing fee of **\$130.00** for furnishing the English translation later than ☐ 20 ☐ 30  
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$

0.00

**TOTAL NATIONAL FEE =**

\$ 890.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be  
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **\$40.00** per property +

\$

40.00

**TOTAL FEES ENCLOSED =**

\$ 930.00

Amount to be refunded:

\$

charged:

\$

a. ☐ A check in the amount of \$ \_\_\_\_\_ to cover the above fees is enclosed.

b. ☒ Please charge my Deposit Account No. 13-3848 in the amount of \$ 930.00 to cover the above fees.  
A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any  
overpayment to Deposit Account No. 13-3848. A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card  
information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

**NOTE:** Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR  
1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



**00157**

PATENT TRADEMARK OFFICE

SIGNATURE

Aron Preis

NAME

29,426

REGISTRATION NUMBER

10/031502  
531 Rec'd PCT/EP 15 JAN 2002

PATENT APPLICATION  
Mo6925  
LeA 33,899

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN APPLICATION OF )  
SILKE KRATSCHEMER ET AL ) PCT/EP00/06377  
SERIAL NO.: TO BE ASSIGNED )  
FILED: HEREWITH )  
TITLE: POLYCARBONATE AND MOLDED )  
POLYCARBONATE ARTICLES )

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents  
Washington, D.C. 20231  
Sir:

Prior to its examination kindly amend the enclosed translation of the captioned patent application as follows.

"Express Mail" mailing label number ET700175893US  
Date of Deposit January 15, 2002

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Assistant Commissioner of Patents and Trademarks, Washington, D.C. 20231

Karen S. Lockhart

(Name of person mailing paper or fee)

[Signature]  
Signature of person mailing paper or fee)

## IN THE SPECIFICATION

On page 1, please delete the title and insert the following in its stead:

--POLYCARBONATE AND MOLDED POLYCARBONATE ARTICLES--

Add the following abstract. A new abstract page is enclosed.

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## POLYCARBONATE AND MOLDED POLYCARBONATE ARTICLES

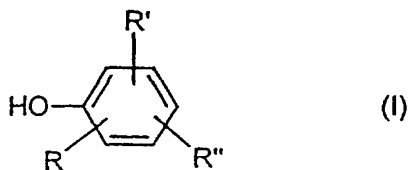
### ABSTRACT OF THE DISCLOSURE

The invention relates to a novel polycarbonate and its use as a material in the production of molded articles and semifinished products, especially for transparent articles such as data memories or audio compact discs, slabs, cellular sheets, films, lamp housings, panes, especially panes for motor vehicles, diffusion glasses, but also for electrical applications or for the construction of houses.--

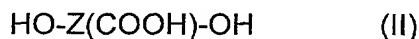
## N THE CLAIMS:

Please amend Claims 1, 3, 4 and 6 as follows:

1. (Amended) Solvent-free, low-branching, thermoplastic, aromatic polycarbonates prepared by the transesterification process and having weight-average molecular weights  $M_w$  of from 2000 to 150,000 based on diphenols, chain terminators of formula (I)



wherein R, R' and R'' each independently of the others represent H, optionally branched  $C_1$ - $C_{34}$ -alkyl/cycloalkyl,  $C_7$ - $C_{34}$ -alkaryl or  $C_6$ - $C_{34}$ -aryl, and, optionally, branching agents, characterised in that after its total saponification it contains less than 300 ppm, determined by HPLC, of elements conforming to formula (II)



wherein Z is an aromatic radical having 6 to 30 carbon atoms and where the acid group is in the ortho position relative to a hydroxy group.

3. (Amended) The polycarbonate according to Claim 1 wherein said elements are present in an amount of from 0.03 ppm to 250 ppm.

4. (Amended) A process for the preparation of the low-branching polycarbonates according to Claim 1 comprising transesterification in the presence of phosphonium salt catalyst.

6. (Amended) The process according to Claim 4 wherein the catalyst is tetraphenylphosphonium phenolate.

Please cancel Claim 7.

Please add new Claims 8 through 10.

--8. A molded article comprising the polycarbonate of Claim 1.

9. The molded article of Claim 8 characterized in that it is transparent.

10. The article of Claim 8 selected from the group consisting of data stores, audio compact disks, sheets, multi-wall sheets, films, lamp housings, panes, headlamp lenses, and structural elements. --

**REMARKS**

The present amendment seeks to place the application in better conformance with U.S. practice. Entry of the amendment is requested.

Respectfully submitted,

By



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Attorney for Applicants  
Reg. No. 29,426

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412-777-8363

s/rmc/ap0044

**VERSION WITH MARKINGS TO SHOW CHANGES MADE**

**IN THE SPECIFICATION**

On page 1, please delete the title and insert the following in its stead:

[POLYCARBONATE AND MOULDED BODIES THEREOF]

POLYCARBONATE AND MOLDED POLYCARBONATE ARTICLES

The following abstract has been added and the abstract page is attached.

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POLYCARBONATE AND MOLDED POLYCARBONATE ARTICLES

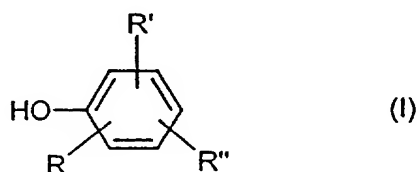
**ABSTRACT OF THE DISCLOSURE**

The invention relates to a novel polycarbonate and its use as a material in the production of molded articles and semifinished products, especially for transparent articles such as data memories or audio compact discs, slabs, cellular sheets, films, lamp housings, panes, especially panes for motor vehicles, diffusion glasses, but also for electrical applications or for the construction of houses.--

**IN THE CLAIMS:**

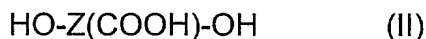
Claims 1, 3, 4 and 6 have been amended as follows:

1. (Amended) Solvent-free, low-branching, thermoplastic, aromatic polycarbonates prepared by the transesterification process and having weight-average molecular weights  $M_w$  of from 2000 to 150,000 [, preferably from 4500 to 55,000,] based on diphenols, chain terminators of formula (I)



wherein R, R' and R'' [may] each independently of the others represent H, optionally branched C<sub>1</sub>-C<sub>34</sub>-alkyl/cycloalkyl, C<sub>7</sub>-C<sub>34</sub>-alkaryl or C<sub>6</sub>-C<sub>34</sub>-aryl, and, optionally, branching agents, characterised in that after its total saponification it contains less

than 300 ppm, determined by HPLC, of elements conforming to [structural elements of] formula (II)



[have a value after total saponification and HPLC determination of less than 300 ppm, Z being as defined for formula (VI) and] wherein Z is an aromatic radical having 6 to 30 carbon atoms and where the acid group is[being] in the ortho position relative to a hydroxy group.

3. (Amended) [Polycarbonates] The polycarbonate according to [one of the above-mentioned claims, characterised in that structural elements of formula (II) have a value after total saponification and HPLC determination] Claim 1 wherein said elements are present in an amount of from 0.03 ppm to 250 ppm.

4. (Amended) [Process] A process for the preparation of the low-branching polycarbonates according to ]the invention, ] Claim 1 comprising transesterification in the presence of [characterised in that phosphonium salts are used as catalyst ] phosphonium salt catalyst.

6. (Amended) [ Process] The process according to Claim 4 [or 5, characterised in that] wherein the catalyst is tetraphenylphosphonium phenolate.

Claim 7 has been cancelled.

Claims 8, 9 and 10 are new and have been added.



POLYCARBONATE AND MOLDED POLYCARBONATE ARTICLES

ABSTRACT OF THE DISCLOSURE

The invention relates to a novel polycarbonate and its use as a material in the production of molded articles and semifinished products, especially for transparent articles such as data memories or audio compact discs, slabs, cellular sheets, films, lamp housings, panes, especially panes for motor vehicles, diffusion glasses, but also for electrical applications or for the construction of houses.

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LeA 33,899

Polycarbonate and moulded bodies thereof

5 The present invention relates to a novel polycarbonate and its use as a material for the production of moulded bodies and semi-finished products, especially for transparent applications, such as data stores or audio compact disks, sheets, multi-wall sheets, films, lamp housings, panes, especially panes for motor vehicles, headlamp lenses, but also for electrical applications or house building.

10 The preparation of aromatic polycarbonates by the melt transesterification process is known from the literature and is described, for example, in the Encyclopedia of Polymer Science, Vol. 10 (1969), Chemistry and Physics of Polycarbonates, Polymer Reviews, H. Schnell, Vol. 9, John Wiley and Sons, Inc. (1964) and, starting from DE 1 031 512, in some patents.

15 In EP-B-360 578 there are described polycarbonates containing end groups other than phenol. The polycarbonates obtained according to EP 360 578 have a markedly increased content of erroneous structures as compared with the polycarbonates synthesised in solution. As a result, such materials have disadvantages as regards melt stability, thermostability and constancy of colour.

20 The object was, therefore, to develop polycarbonates and a process for their preparation, having greater stability.

The object has been achieved by the synthesis of polycarbonates having a markedly reduced concentration of erroneous structures.

The present invention provides solvent-free, low-branching, thermoplastic, aromatic polycarbonates prepared by the transesterification process and having weight-average molecular weights  $M_w$  of from 2000 to 150,000, preferably from 4500 to 55,000, based on diphenols, chain terminators of formula (I)

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"Express Mail" mailing label number PT70017589345Date of Deposit January 15, 2002

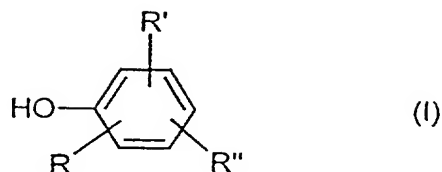
I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Assistant Commissioner of Patents and Trademarks, Washington, D.C. 20231

Karen S. Lockhart

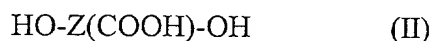
(Name of person mailing paper or fee)

Signature of person mailing paper or fee

- 2 -



wherein R, R' and R'' may each independently of the others represent H, optionally  
 branched C<sub>1</sub>-C<sub>34</sub>-alkyl/cycloalkyl, C<sub>7</sub>-C<sub>34</sub>-alkaryl or C<sub>6</sub>-C<sub>34</sub>-aryl, and, optionally,  
 5 branching agents, characterised in that structural elements of formula (II)

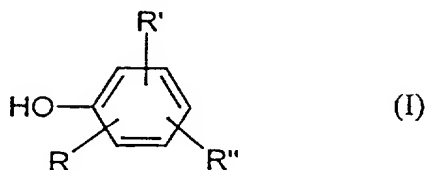


have a value after total saponification and HPLC determination of less than  
 10 300 ppm, preferably from 0.03 ppm to 250 ppm, Z being as defined for formula (VI)  
 and the acid group being in the ortho position relative to a hydroxy group.

As compared with polycarbonates mis-structured in the usual manner, the  
 polycarbonates according to the invention exhibit a substantially increased  
 15 hydrolytic stability and an improved critical thickness, with otherwise comparable  
 mechanical and thermal properties.

The polycarbonates according to the invention are prepared in the melt from  
 dihydroxy compounds, dicarbonates, chain terminators and, optionally, branching  
 20 agents.

Chain terminators within the context of the invention are those of formula (I)

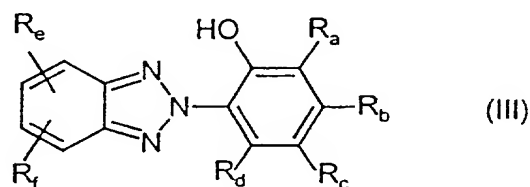


- 3 -

wherein R, R' and R" may each independently of the others represent H, optionally branched C<sub>1</sub>-C<sub>34</sub>-alkyl/cycloalkyl, C<sub>7</sub>-C<sub>34</sub>-alkaryl or C<sub>6</sub>-C<sub>34</sub>-aryl, for example

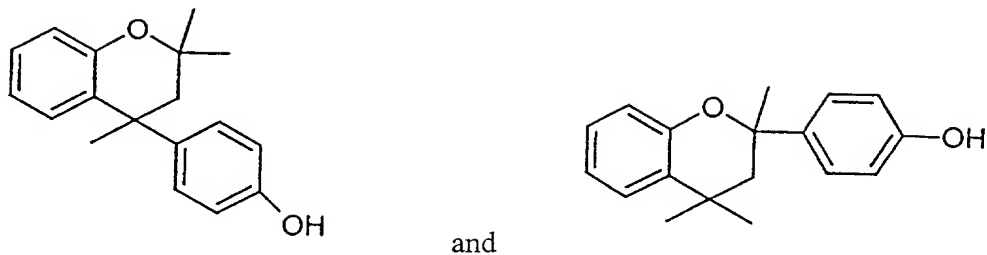
o-n-butylphenol, m-n-butylphenol, p-n-butylphenol,  
 5 o-isobutylphenol, m-isobutylphenol, p-isobutylphenol,  
 o-tert-butylphenol, m-tert-butylphenol, p-tert-butylphenol,  
 o-n-pentylphenol, m-n-pentylphenol, p-n-pentylphenol,  
 o-n-hexylphenol, m-n-hexylphenol, p-n-hexylphenol,  
 o-cyclohexylphenol, m-cyclohexylphenol, p-cyclohexylphenol,  
 10 o-phenylphenol, m-phenylphenol, p-phenylphenol,  
 o-isooctylphenol, m-isooctylphenol, p-isooctylphenol,  
 o-n-nonylphenol, m-n-nonylphenol, p-n-nonylphenol,  
 o-cumylphenol, m-cumylphenol, p-cumylphenol,  
 o-naphthylphenol, m-naphthylphenol, p-naphthylphenol,  
 15 2,5-di-tert-butylphenol, 2,4-di-tert-butylphenol, 3,5-di-tert-butylphenol,  
 2,5-dicumylphenol, 3,5-dicumylphenol,  
 4-phenoxyphenol, 2-phenoxyphenol, 3-phenoxyphenol,  
 3-pentadecylphenol, 2-pentadecylphenol, 4-pentadecylphenol,  
 2-phenylphenol, 3-phenylphenol, 4-phenylphenol,  
 20 tritylphenol, 3-triphenylmethylphenol, 2-triphenylmethylphenol,

also benzotriazole derivatives of the general formula (III)



25 wherein R<sub>a</sub> to R<sub>f</sub> are as defined above for R, R' and R",  
 and chroman compounds such as

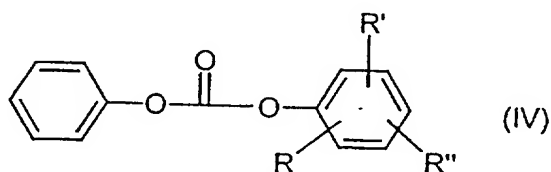
- 4 -



preferably high-boiling phenols such as tritylphenol, cumylphenol, pentadecylphenol or chromans,

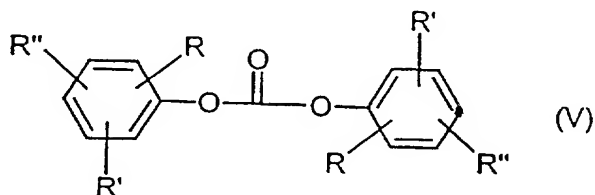
5

or also in the form of compounds that are capable of transesterification under the synthesis conditions, such as, for example, carbonates, oxalates, o-carboxylic acid esters or the like, with preference being given to the free phenols or the carbonic acid diesters of formula (IV)



10

and formula (V)



15 wherein R, R' and R'' correspond to those of formula (I). Phenols or transesterification-active substances can be added to the synthesis individually or in the form of a mixture. Preferred mixtures are those with diphenyl carbonate. It is possible to add the phenol or the phenol-carrying compound at any time during the reaction, preferably at the beginning of the reaction, and the addition can be divided

into several portions. The total amount of carbonic acid ester is from 100 to 130 mol%, preferably from 103 to 120 mol%, based on the dihydroxy compound.

For the preparation of polycarbonates by the process according to the invention it is possible to use a chain terminator or a mixture of several chain terminators, so that there may be present as an end group in the polycarbonate according to the invention phenol but also phenol together with other chain terminators. From 0.4 to 17 mol%, particularly preferably from 1.3 to 8.6 mol% (based on the dihydroxy compound), of chain terminator are preferably added. The addition may take place either before the reaction or wholly or partially during the reaction.

Dihydroxy compounds within the context of the invention are those of formula (VI)



in which Z is an aromatic radical having from 6 to 30 carbon atoms which may contain one or more aromatic nuclei, may be substituted and may contain aliphatic or cycloaliphatic radicals or alkylaryls or hetero atoms as bridge members.

Examples of dihydroxy compounds of formula (VI) are

hydroquinone,

resorcinol,

dihydroxydiphenyls,

bis-(hydroxyphenyl)-alkanes,

bis-(hydroxyphenyl)-cycloalkanes,

bis-(hydroxyphenyl) sulfides,

bis-(hydroxyphenyl) ethers,

bis-(hydroxyphenyl) ketones,

bis-(hydroxyphenyl)-sulfones,

bis-(hydroxyphenyl) sulfoxides,

$\alpha,\alpha'$ -bis-(hydroxyphenyl)-diisopropylbenzenes  
and their nuclear-alkylated and nuclear-hydrogenated compounds.

These and other suitable diphenols are described, for example, in US-A 3 028 365, 3  
148 172, 3 275 601, 2 991 273, 3 271 367, 3 062 781, 2 970 131 and 2 999 846, in  
German Offenlegungsschrift 1 570 703, 2 063 050, 2 063 052, 2 211 0956, in French  
Patent Specification 1 561 518 and in the monograph "H. Schnell, Chemistry and  
Physics of Polycarbonates, Interscience Publishers, New York 1964".

- 10 Examples of preferred diphenols are:
- 4,4'-dihydroxydiphenyl,
  - 2,2-bis-(4-hydroxyphenyl)propane,
  - 2,4-bis-(4-hydroxyphenyl)-2-methylbutane,
  - 1,1-bis-(4-hydroxyphenyl)cyclohexane,
  - 15 1,1-bis-(4-hydroxyphenyl)-4-methylcyclohexane,
  - $\alpha,\alpha'$ -bis-(4-hydroxyphenyl)-p-diisopropylbenzene,
  - $\alpha,\alpha'$ -bis-(4-hydroxyphenyl)-m-diisopropylbenzene,
  - bis-(4-hydroxyphenyl)sulfone,
  - bis-(4-hydroxyphenyl)methane,
  - 20 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane,
  - 2,2-bis-(2,6-dimethyl-4-hydroxyphenyl)propane,
  - 2,2-bis-(4-hydroxyphenyl)hexafluoropropane,
  - 1,1-(4-hydroxyphenyl)-1-phenylethane,
  - bis-(4-hydroxyphenyl)diphenylmethane,
  - 25 dihydroxydiphenyl ether,
  - 4,4'-thiobisphenol,
  - 1,1-bis-(4-hydroxyphenyl)-1-(1-naphthyl)ethane,
  - 1,1-bis-(4-hydroxyphenyl)-1-(2-naphthyl)ethane,
  - 2,3-dihydroxy-3-(4-hydroxyphenyl)-1,1,3-trimethyl-1*H*-inden-5-ol,
  - 30 2,3-dihydroxy-1-(4-hydroxyphenyl)-1,3,3-trimethyl-1*H*-inden-5-ol,

2,2',3,3'-tetrahydro-3,3',3'-tetramethyl-1,1'-spirobi[1*H*-indene]-5,5'-diol.

Special preference is given to  
resorcinol,

- 5 1,1-bis-(4-hydroxyphenyl)-1-(1-naphthyl)ethane,  
1,1-bis-(4-hydroxyphenyl)-1-(2-naphthyl)ethane,  
2,2-bis-(4-hydroxyphenyl)propane,  
 $\alpha,\alpha'$ -bis-(4-hydroxyphenyl)-p-diisopropylbenzene,  
 $\alpha,\alpha'$ -bis-(4-hydroxyphenyl)-m-diisopropylbenzene,  
10 1,1-bis-(4-hydroxyphenyl)cyclohexane,  
1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane,  
bis-(4-hydroxyphenyl)diphenylmethane,  
4,4'-dihydroxydiphenyl.

- 15 It is possible to use both one diphenol of formula (VI), with formation of homo-  
polycarbonates, and several diphenols of formula (VI), with formation of  
copolycarbonates.

- 20 Low-branching within the context of the invention means that the content of formula  
(II) in the polycarbonate has a value after total saponification and HPLC  
determination of less than 300 ppm, preferably from 0.03 ppm to 250 ppm.

- 25 The polycarbonates can be branched in a deliberate and controlled manner by the use  
of small amounts of from 0.02 to 3.6 mol% (based on the dihydroxy compound) of  
branching agents. Suitable branching agents are the compounds which are suitable  
for the preparation of polycarbonates and contain three or more functional groups,  
preferably those containing three or more than three phenolic OH groups.

- 30 Examples of suitable branching agents are  
phloroglucinol,

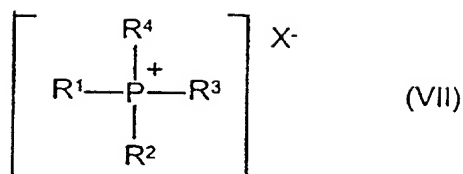


- 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)heptane,  
 1,3,5-tri-(4-hydroxyphenyl)benzene,  
 1,1,1-tri-(4-hydroxyphenyl)ethane,  
 tri-(4-hydroxyphenyl)phenylmethane,  
 5 2,2-bis-[4,4-bis(4-hydroxyphenyl)cyclohexyl]propane,  
 2,4-bis-(4-hydroxyphenyl-isopropyl)phenol,  
 2,6-bis-(2-hydroxy-5-methylbenzyl)-4-methylphenol,  
 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)propane,  
 hexa-[4-(4-hydroxyphenyl-isopropyl)phenyl]orthoterephthalic acid ester,  
 10 tetra-(4-hydroxyphenyl)methane,  
 tetra-[4-(4-hydroxyphenyl-isopropyl)phenoxy]methane,  
 1,4-bis-[4',4''-dihydroxytriphenyl)methyl]benzene,  
 $\alpha,\alpha',\alpha''$ -tris-(4-hydroxyphenyl)-1,3,4-triisopropenylbenzene,  
 isatinbiscresol,  
 15 pentaerythritol,  
 2,4-dihydroxybenzoic acid,  
 trimesic acid,  
 cyanuric acid.
- 20 1,1,1-tri-(4-hydroxyphenyl)ethane and isatinbiscresol are particularly preferred.

As catalyst for the preparation of the polycarbonates according to the invention there may be used phosphonium salts, optionally in combination with other suitable catalysts which do not lead to erroneous structures such as formula (II), such as, for  
 25 example, other onium compounds.

Phosphonium salts within the context of the invention are those of formula (VII)

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wherein  $R^{1-4}$  may be the same or different  $C_1$ - $C_{18}$ -alkyls,  $C_6$ - $C_{14}$ -aryls,  $C_7$ - $C_{12}$ -aralkyls or  $C_5$ - $C_6$ -cycloalkyls, preferably methyl or  $C_6$ - $C_{14}$ -aryls, particularly preferably methyl or phenyl, and  $X^-$  may be an anion, such as a sulfate, hydrogen sulfate, hydrogen carbonate, carbonate, acetate, boranate, hydrogen phosphates, a halide, preferably a fluoride, chloride or bromide, or an alcoholate of the formula OR wherein R may be  $C_6$ - $C_{14}$ -aryl or  $C_7$ - $C_{12}$ -aralkyl, preferably phenyl.

Preferred catalysts are  
tetraphenylphosphonium fluoride,  
tetraphenylphosphonium tetraphenylboranate,  
particularly preferably tetraphenylphosphonium phenolate.

The preparation of the polycarbonates according to the invention is carried out, for example, as follows: in the first step, the diphenols, the carbonic acid diesters, the catalyst and, optionally, the alkylphenols and branching agents are melted at temperatures of from  $75^\circ\text{C}$  to  $225^\circ\text{C}$ , preferably from  $105^\circ\text{C}$  to  $235^\circ\text{C}$ , particularly preferably from  $120^\circ\text{C}$  to  $190^\circ\text{C}$ , under normal pressure within a period of from 0.1 to 5 hours, preferably from 0.25 to 3 hours. Then the oligocarbonate is prepared by applying a vacuum and raising the temperature by removing the monophenol by distillation. In the last step, the polycarbonate is prepared in the polycondensation by raising the temperature further to from  $240^\circ\text{C}$  to  $325^\circ\text{C}$  and at a pressure of  $< 2$  mbar.

25

In the preparation of polycarbonates by the melt transesterification process, the reaction of the bisphenol and the carbonic acid diester may be carried out

continuously or discontinuously, for example, in stirrer vessels, thin-layer evaporators, falling film evaporators, stirrer vessel cascades, extruders, kneaders, simple disk reactors and high-viscosity disk reactors.

- 5 Isolation of the polycarbonates according to the invention is likewise carried out in a known manner, for example by removing, spinning and granulating.

10 The polycarbonates according to the invention can have weight-average molecular weights  $M_w$  of approximately from 2000 to 150,000, preferably approximately from 4500 to 55,000,  $M_w$  being determined *via* the relative solution viscosity in dichloromethane or in mixtures of equal amounts by weight of phenol/o-dichlorobenzene, calibration being by means of light scattering.

15 The polycarbonates according to the invention have the usual OH end group contents known from the literature, which can be determined by photometry using titanium tetrachloride.

20 The polycarbonates according to the invention can be processed thermoplastically in the usual manner at temperatures of from 260°C to 320°C. Moulded bodies and films of any kind can be produced in a known manner by injection moulding or by means of extrusion.

25 The polycarbonates according to the invention are readily soluble in solvents such as chlorinated hydrocarbons, for example methylene chloride, and can therefore be processed, for example, in a known manner to cast films.

Accordingly, the invention also provides a process for the preparation of the low-branching polycarbonates according to the invention, characterised in that there are used as catalyst phosphonium salts of formula (VII) in concentrations of from  
30  $10^{-2}$  mol to  $10^{-6}$  mol, based on 1 mol of diphenol, optionally in combination with

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other suitable catalysts which do not lead to erroneous structures such as formula (II), such as, for example, other onium compounds.

5 In order to improve the properties, auxiliary substances and reinforcing agents can be added to the polycarbonates according to the invention. The following, *inter alia*, are to be taken into consideration as such: stabilisers, flow auxiliaries, mould-release agents, fireproofing agents, pigments, finely divided minerals, fibrous materials, for example alkyl and aryl phosphites, phosphates and phosphanes, low-molecular-weight carboxylic acid esters, halogen compounds, salts, chalk, quartz powder, glass  
10 and carbon fibres, pigments and combinations thereof.

Other polymers, for example polyolefins, polyurethanes, polyesters and polystyrene, may also be added to the polycarbonates according to the invention.

15 Excellent properties are achieved with that material even in moulded bodies.

Accordingly, the present invention relates also to the use of the low-branching, solvent-free, aromatic polycarbonates according to the invention for the production of moulded bodies and semi-finished products, especially for transparent  
20 applications, such as data stores or audio compact disks, sheets, multi-wall sheets, films, lamp housings, panes, especially panes for motor vehicles, headlamp lenses, but also for electrical applications or house building.

## Examples

### Comparison Example 1

5 45.60 g (0.2 mol) of bisphenol A, 47.08 g (110 mol%, based on bisphenol A) of diphenyl carbonate, 3.7 mg (0.03 mol%, based on bisphenol A) of boric acid and 2.12 g (5 mol%, based on bisphenol A) of 4-cumylphenol are weighed into a 500 ml three-necked flask equipped with a stirrer, an internal thermometer and a Vigreux column (30 cm, mirrored) with a bridge. The apparatus is freed of atmospheric  
10 oxygen by applying a vacuum and flushing with nitrogen (three times) and the mixture is melted at 180°C and stirred for 30 minutes. Then 36.5 mg (0.03 mol%, based on bisphenol A) of a 15% ammonium hydroxide solution and 0.5 mg (0.003 mol%, based on bisphenol A) of sodium hydrogen carbonate are added and stirring is continued for a further 30 minutes. The temperature is raised to 210°C and the  
15 vacuum is increased to 200 mbar and the phenol that forms is removed by distillation. After one hour, the temperature is raised to 240°C and after 20 minutes the vacuum is reduced to 150 mbar. After a further 20 minutes, the pressure is lowered to 100 mbar and maintained for 20 minutes. Then the pressure is reduced to 15 mbar for 30 minutes. The temperature is then raised to 270°C, the vacuum is  
20 reduced to 0.5 mbar and stirring is carried out for a further 2 hours. The results are summarised in Table 1.

### Example 1

25 As Comparison Example 1, but 4.9 mg (0.004 mol%, based on bisphenol A) of tetraphenylphosphonium phenolate (which is metered in in the form of mixed crystal containing 30 wt.% phenol, based on the mixed crystal) are added instead of tetramethylammonium hydroxide. Sodium hydrogen carbonate and boric acid are not added. The results are summarised in Table 1.

**Example 2**

45.66 g (0.2 mol) of bisphenol A, 47.13 g (110 mol%, based on bisphenol A) of diphenyl carbonate, 4.9 mg (0.004 mol%, based on bisphenol A) of tetraphenylphosphonium phenolate (which is metered in in the form of mixed crystal containing 30 wt.% phenol, based on the mixed crystal) and 2.12 g (5 mol%, based on bisphenol A) of 4-cumylphenol are weighed into a 500 ml three-necked flask equipped with a stirrer, an internal thermometer and a Vigreux column (30 cm, mirrored) with a bridge. The apparatus is freed of atmospheric oxygen by applying a vacuum and flushing with nitrogen (three times) and the mixture is melted at 150°C. The temperature is raised to 190°C and the vacuum is increased to 100 mbar and the phenol that forms is removed by distillation. After 20 minutes, the temperature is raised to 235°C and the vacuum is reduced to 60 mbar. After 15 minutes, the temperature is raised to 250°C and after a further 15 minutes the vacuum is increased to 5 mbar. The mixture is then heated to 280°C and the pressure is reduced to 0.5 mbar after 15 minutes. After a further 15 minutes, the mixture is stirred at 300°C for a further 30 minutes. The results are summarised in Table 1.

**Example 3**

45.66 g (0.2 mol) of bisphenol A, 47.13 g (110 mol%, based on bisphenol A) of diphenyl carbonate, 4.9 mg (0.004 mol%, based on bisphenol A) of tetraphenylphosphonium phenolate (which is metered in in the form of mixed crystal containing 30 wt.% phenol, based on the mixed crystal) and 3.05 g (5 mol%, based on bisphenol A) of 3-pentadecylphenol are weighed into a 500 ml three-necked flask equipped with a stirrer, an internal thermometer and a Vigreux column (30 cm, mirrored) with a bridge. The apparatus is freed of atmospheric oxygen by applying a vacuum and flushing with nitrogen (three times) and the mixture is melted at 150°C. The temperature is raised to 190°C and the vacuum is increased to 100 mbar and the phenol that forms is removed by distillation. After 30 minutes, the temperature is

raised to 235°C, and after a further 30 minutes it is increased to 300°C. The vacuum is slowly reduced to 0.5 mbar and the mixture is stirred for a further 30 minutes. The results are summarised in Table 1.

#### 5 Example 4

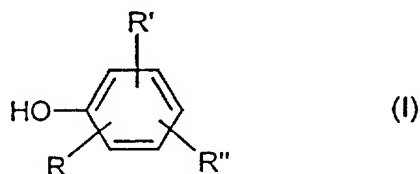
1141.47 g (5 mol) of bisphenol A, 1113.94 g (104 mol%, based on bisphenol A) of diphenyl carbonate and 122.5 mg (0.004 mol%, based on bisphenol A) of tetraphenylphosphonium phenolate (which is metered in in the form of mixed crystal containing 30 wt.% phenol, based on the mixed crystal) are weighed into a stirred container. The container is freed of atmospheric oxygen by applying a vacuum and flushing with nitrogen (three times) and the mixture is melted at 150°C. The temperature is raised to 190°C and the vacuum is increased to 100 mbar and the phenol that forms is removed by distillation. After 60 minutes, the temperature is raised to 235°C, and after a further 30 minutes the vacuum is slowly reduced to 60 mbar and stirring is carried out for a further 15 minutes. The mixture is then heated to 250°C and after 15 minutes the pressure is lowered to 5 mbar for a short time. The results are summarised in Table 1.

20 Table 1

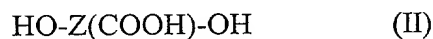
	Chain terminator	Formula (II) [ppm]	Solution viscosity
Comparison Example 1	cumylphenol/phenol	303	1.142
Example 1	cumylphenol/phenol	70	1.160
Example 2	cumylphenol/phenol	25	1.122
Example 3	pentadecylphenol/phenol	86	1.161
Example 4	phenol	12	1.124

**Patent Claims**

1. Solvent-free, low-branching, thermoplastic, aromatic polycarbonates prepared by the transesterification process and having weight-average molecular weights  $M_w$  of from 2000 to 150,000, preferably from 4500 to 55,000, based on diphenols, chain terminators of formula (I)



- wherein R, R' and R'' may each independently of the others represent H, optionally branched  $C_1$ - $C_{34}$ -alkyl/cycloalkyl,  $C_7$ - $C_{34}$ -alkaryl or  $C_6$ - $C_{34}$ -aryl, and, optionally, branching agents, characterised in that structural elements of formula (II)



- have a value after total saponification and HPLC determination of less than 300 ppm, Z being as defined for formula (VI) and the acid group being in the ortho position relative to a hydroxy group.

2. Polycarbonates according to claim 1, characterised in that the end groups consist of alkylphenol end groups to the extent of more than 30% of the reacted end groups.
3. Polycarbonates according to one of the above-mentioned claims, characterised in that structural elements of formula (II) have a value after total saponification and HPLC determination of from 0.03 ppm to 250 ppm.



4. Process for the preparation of the low-branching polycarbonates according to the invention, characterised in that phosphonium salts are used as catalyst.
5. Process according to claim 4, in which the catalyst is used in concentrations of from  $10^{-2}$  mol to  $10^{-6}$  mol, based on 1 mol of diphenol.
6. Process according to claim 4 or 5, characterised in that the catalyst is tetraphenylphosphonium phenolate.
- 10 7. Use of the low-branching, solvent-free, aromatic polycarbonates according to the invention for the production of moulded bodies and semi-finished products, especially for transparent applications, such as data stores or audio compact disks, sheets, multi-wall sheets, films, lamp housings, panes, especially panes for motor vehicles, headlamp lenses, but also for electrical  
15 applications or house building.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought

on the invention entitled

**POLYCARBONATE AND MOLDED POLYCARBONATE ARTICLES**

the specification of which is attached hereto,

or was filed on **July 6, 2000**

as a PCT Application Serial No. **PCT/EP00/06377**

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s), the priority(ies) of which is/are to be claimed:

**199 33 128.6**  
(Number)

**Germany**  
(Country)

**July 19, 1999**  
(Month/Day/Year Filed)

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose the material information as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)

(Filing Date)

(Status)

(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)

(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Le A 33 899-US

OK

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